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Review of Distribution Coefficients for Radionuclides in Carbonate Minerals

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Introduction

An understanding of the transport of radionuclides in carbonate minerals is necessary to be able to predict the fate of (and potentially remediate) radionuclides in the environment. In some environments, carbonate minerals such as calcite, aragonite, dolomite and limestone are present and an understanding of the sorption of radionuclides in these carbonate minerals is therefore advantageous. A list of the radionuclides of interest is given in Table 1.

The distribution coefficient, K_d is defined as the ratio of the contaminant concentration bound on the solid phase to the contaminant concentration remaining in the liquid phase at equilibrium. Some authors report distribution coefficients and others report partition coefficients, the data presented in this work assumes equality between these two terms, and data are presented and summarized in this work as logarithmic distribution coefficients (log K_d).

Published literature was searched using two methods. Firstly, the JNC Sorption Database, namely Shubutani et al (1999), and Suyama and Sasamoto (2004) was used to select elements of interest and a number of carbonate minerals. Secondly, on-line literature search tools were used to locate relevant published articles from 1900 to 2009.

Over 300 data points covering 16 elements (hydrogen, carbon, calcium, nickel, strontium, technetium, palladium, iodine, cesium, samarium, europium, holmium, uranium, neptunium, plutonium and americium) were used to calculate an average and range of log K_d values for each element. Unfortunately, no data could be found for chlorine, argon, krypton, zirconium, niobium, tin, thorium and curium. A description of the data is given below, together with the average, standard deviation, minimum, maximum and number of inputs for radionuclide K_d values for calcite, aragonate, limestone, dolomite and unidentified carbonate rocks in Table 2. Finally, the data are condensed into one group (carbonate minerals) of data for each element of interest in Table 3.

Table 1. Elements and radionuclides of interest in studying subsurface migration of radioactivity in the environment.

Element	Radionuclides of Interest
Hydrogen	^{3}H
Carbon	^{14}C
Chlorine	^{36}Cl
Argon	^{39}Ar
Calcium	^{41}Ca
Nickel	^{59}Ni, ^{63}Ni
Krypton	^{85}Kr
Strontium	^{90}Sr
Zirconium	^{93}Zr
Niobium	^{93m}Nb, ^{94}Nb
Technetium	^{99}Tc
Palladium	^{107}Pd
Tin	^{121m}Sn, ^{126}Sn
Iodine	^{129}I
Cesium	^{135}Cs, ^{137}Cs
Samarium	^{151}Sm
Europium	^{150}Eu, ^{152}Eu, ^{154}Eu
Holmium	^{166}Ho
Thorium	^{232}Th
Uranium	^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U
Neptunium	^{237}Np
Plutonium	^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu
Americium	^{241}Am, ^{243}Am
Curium	^{244}Cm

Hydrogen

Hydrogen (in particular, tritium) has long been used as a conservative tracer in fate and transport experiments. A review by the Environmental Protection Agency (1999) supported the use of a zero K_d value.

Carbon

Sheppard et al (1998) studied the sorption of ^{14}C on calcite, montmorillonite and soil. The partition coefficient of three size fractions of natural calcite and one of synthetic calcite were measured, yielding log K_d values ranging from 0.90 to 1.70 for natural calcite and 1.93 for synthetic calcite. Additionally, Reimus et al (2008) noted sorption of carbon on calcite and dolomite with log K_d 2.30 and 1.40 respectively.

Chlorine

No data were available for chlorine transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Argon
No data were available for argon transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Calcium
Only one source was found for calcium sorption on carbonate minerals. Zavarin et al (2004) proposes log K_d values ranging from 0.44 and 0.52 for Frenchman Flat and Pahute Mesa calcite systems respectively.

Nickel
Zachara et al (1991) measured exchange constants of divalent metal ions on calcite. Their work yielded log K_d of 0.51 for nickel sorption on calcite. The JNC database lists only one reference for nickel on carbonate minerals, namely log K_d -2.24 for Ni on calcite, derived by Cornell and Aksoyoglu (1992) at pH 7.2 under nitrogen.

Krypton
No data were available for krypton transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Strontium
Andersson et al (1983a) measured strontium sorption on calcite at pH 6 to 9 and found log K_d values ranging from -2.70 to -2.40, while Fujikawa and Fukui (1997) measured log K_d at -2.54 to -0.60 from pH 4.8 to 10.1. Torstenfelt et al (1981) measured sorption at pH 8 to 9 and found log K_d -3.00 to -2.15 and Ticknor et al (1991) measured log K_d for Sr on calcite at -2.52 with no identified pH. Curti (1999) summarizes the work of Lorens (1981), Katz et al (1972), Mucci and Morse (1983), Pingitore and Eastman (1986), and Tesoriero and Pankow (1996) with a range of log K_d values for Sr on calcite from -1.52 to -0.57. Zachara et al (1991) measured exchange constants of divalent metal ions on calcite. Their work yielded log K_d of -2.04 for strontium sorption on calcite. Zavarin et al (2004) proposes log K_d values ranging from -1.31 to -1.24 for Frenchman Flat and Pahute Mesa calcite respectively. Maclean et al (1978) measured strontium sorption on limestone at pH 8.2 and found log K_d -2.57, while Relyea et al (1978) measured sorption at pH 7 to 8.2 and found log K_d ranging from -2.62 to 1.86 for Sr. Andersson et al (1983b) provided a literature review of radionuclide sorption on geologic media between pH 7 and 9, which included data from Seitz et al (1979), Meyer (1978), Maclean et al (1978), Relyea et al (1978), Serne et al (1977), Serne et al (1979), Ramspott et al (1977), and Furnica et al (1973) for strontium sorption on limestone. Values for log K_d ranged from -3.0 to 1.0. Andersson et al (1983a) measured strontium sorption on dolomite at pH 8 to 8.5, yielding log K_d -2.52 and Maclean et al (1978) found log K_d equal to -3.00 at pH 7.7. Andersson's literature review (1983b) included data from Dosch (1978a), Dosch and Lynch (1978b), Dosch 1979, Dosch and Lynch 1979b, and Hinkenbein and Hlava (1977), Borg et al (1976), Maclean et al (1978), Serne et al (1977) and Miettinen et al (1981) for strontium sorption on dolomite. Values for log K_d ranged from -3.0 to -0.74. Hu et al (2008) measured strontium sorption on

carbonate mineral in the presence of LCA water, with log K_d ranging from -0.19 to 0.11.

Zirconium
No data were available for zirconium transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Niobium
No data were available for niobium transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Technetium
Relyea et al (1978) measured technetium sorption on limestone with log K_d values ranging from -2.94 to -1.11. Additionally, Ito and Kanno (1988) reported Tc sorption on limestone with log K_d -3.05 and -1.76 for oxidizing and reducing conditions respectively. Andersson's literature review (1983b) contained data for technetium sorption on limestone, including references to Strickert et al (1980), Eichholz (1980) and Serne et al (1977), with log K_d values ranging from -3.00 to -2.15. Simiarly, the review included data on Tc sorption on dolomite, with references to Dosch (1979), Strickert et al (1980) and Serne et al (1977), with log K_d values ranging from -3.70 to -1.82. Recent UGTA data from Hu et al (2008) suggest the sorption of technetium on carbonate rocks in synthetic LCA water under atmospheric conditions ranged from log K_d -0.92 to -0.44.

Palladium
Only one reference to palladium sorption on carbonate minerals was found. Dikikh et al (2007) derived log K_d 2.28 for Pd on calcite.

Tin
No data were available for tin transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Iodine
Andersson's literature review (1983b) included data for iodine sorption on limestone with references to Meyer (1979) and Strickert et al (1980) and a log K_d of -1.80. Similarly, the review contained data for iodine on dolomite, with data from Meyer (1979), Dosch (1978a), Fried et al (1978) and Strickert et al (1980) yielding a log K_d value of -3.0. Hu et al (2008) documents log Kd for iodine on carbonate rocks in synthetic LCA water under atmospheric conditions as being from -0.16 to -0.12.

Cesium
Fujikawa and Fukui (1997) measured cesium sorption on calcite at pH 4.5 to 10.1 and reported log K_d from -3.57 to -2.21. Andersson et al (1983a) reported Cs calcite log K_d ranging from -3 to -2.22 at pH 8 to 9. Ticknor et al (1991) also reported measuring log K_d equal to -3 for calcite, although the pH was not apparent. Torstenfelt et al (1981) measured log K_d values ranging from -3 to -2.15 for cesium

on calcite between pH 8 and 9. Maclean et al (1978) reported log K_d values ranging from -3.24 to -1.22 for cesium on limestone between pH 6.5 and 8.2. Relyea et al (1978) measured log K_d between -2.70 to 0.28 at pH 7 to 8.2. Andersson et al (1983a) measured a log K_d for cesium on dolomite of -2.30 at pH 8 to 8.5, while Maclean et al (1978) reported a value of -1.48 at pH 7.7. Andersson's literature review (1983b) included data for cesium sorption on limestone with references to Seitz et al (1978), Maclean et al (1978), Meyer (1978), Relyea et al (1977), Serne et al (1977) and Eichholz (1979) yielding a log K_d range from -1.85 to 0.88. Similarly, the review contained data for cesium sorption on dolomite, with data from Dosch (1978a), Dosch (1979), Lynch and Dosch (1980), Maclean et al (1978), Ramspott (1979), Serne et al (1977) and Eichholz (1979) with log K_d ranging from -3.30 to 0.42. There is one reference in the Andersson review (1983b) to the work of Borg et al (1976) for cesium sorption on carbonate minerals and a log K_d of -1.85. Hu et al (2008) reported UGTA-specific data for cesium sorption on carbonate mineral in equilibrium with LCA water, with a log K_d ranging from -0.6 to -0.36.

Samarium

Zavarin et al (2005) postulated log K_d 3.60 for Sm on calcite. Zhong and Mucci (1995) report similar data for calcite with a log K_d of 3.54. Hull and Schafer (2008) reported log K_d for Sm and calcite equal to 1.04 at pH 7.4. Zavarin et al (2004) proposes log K_d values ranging from 4.46 to 4.67 for Frenchman Flat and Pahute Mesa calcite systems respectively.

Europium

Zhong and Mucci (1995) measured a log K_d of 3.46 for europium sorption on calcite in seawater, which is similar to that of Zavarin et al (2005) with a log K_d of 3.60. Stipp and Lakshtanov (2003), measured the sorption on calcite at pH 6.0 to 6.7 and derived log K_d ranging from 1.09 to 1.97, while later (Lakshtanov and Stipp, 2004) reporting log K_d of 2.89 at pH 6.0 to 6.7. Piriou et al (1997) measured Eu sorption on calcite at pH 8.3, yielding a log K_d of 6.62. Zavarin et al (2004) proposes log Kd values ranging from 4.02 to 4.67 for Frenchman Flat and Pahute Mesa calcite systems respectively.

Holmium

The only available data found on holmium sorption on carbonate minerals was derived by Zhong and Mucci (1995) in seawater at pH 6.8 to 8.3, with a log K_d of 3.06.

Thorium

No data were available for thorium transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Uranium

Ticknor (1993) measured uranium sorption on calcite at pH 7 under various O_2 partial pressures and derived a range of log K_d values from -2.16 to -0.49. Curti (1999) reviewed the work of Meece and Benninger (1993), and Kitano and Oomori

(1971) with log K_d values of -0.66 and -1.40 respectively. Zavarin et al (2004) proposes log K_d values ranging from -3.07 to -1.80 for Frenchman Flat and Pahute Mesa calcite systems respectively. Francis and Bondietti (1979) measured log K_d values for uranium and dolomite at pH 8.6 and reported a range from -2.16 to -1.92. Hu et al (2008) reported UGTA-specific data for uranium sorption on carbonate mineral in equilibrium with LCA water, with a log K_d ranging from 0.20 to 0.45.

Neptunium
A large amount of data is available in a report by Higgo et al (1983), where sorption on calcite materials in seawater at pH 8.2 yields a range of log K_d values for neptunium between 0.08 and 0.93. Stout and Carroll (1993) reviewed data pertaining to several radionuclides (including neptunium) on carbonate minerals. Their review included neptunium calcite sorption research presented by Higgo and Rees (1986), Allard (1984) and Ames and Rai (1978), with log K_d values of 3.6, 2.3 to 3.6, and 3.7 respectively. These values compare to those derived by Keeney-Kennicutt and Morse (1984) and Zavarin et al (2005) of 3.26 and 2.34 respectively. Francis and Bondietti (1979) measured Np sorption on dolomite and reported log K_d values raning from -1.54 to -0.70 at pH 8.6. Zavarin et al (2004) proposes log K_d values ranging from 1.58 and 1.84 for Frenchman Flat and Pahute Mesa calcite systems respectively. Hu et al (2008) reported UGTA-specific data for neptunium sorption on carbonate mineral in equilibrium with LCA water, with a log K_d ranging from 2.33 to 2.47.

Plutonium
Zavarin et al (2005) proposed a log K_d value of 3.04 for Pu(IV) sorption on calcite. Francis and Bondietti (1979) determined a range of log K_d values for Pu(IV) on dolomite at pH 8.6 to be between -0.11 to 0.33. Keeney-Kennicutt and Morse (1985) determined a log Kd for Pu(V) on calcite as 2.74, while Zavarin et al (2005) proposed a log K_d value of 1.30 for Pu(V) sorption on calcite. Zavarin et al (2004) proposes Pu (V) log K_d values ranging from 1.69 and 2.1 for Frenchman Flat and Pahute Mesa calcite systems respectively. Hu et al (2008) reported UGTA-specific data for plutonium sorption on carbonate mineral in equilibrium with LCA water, with a log K_d ranging from 2.03 to 2.09.

Americium
A large amount of data is available in a report by Higgo et al (1983), where sorption on calcite materials in seawater at pH 8.2 yields a range of log K_d values for americium between 1.46 and 2.49. Stout and Carroll (1993) reviewed data pertaining to several radionuclides (including americium) on carbonate minerals. Their review included americium calcite sorption research presented by Shanbhag and Morse (1982), Higgo and Rees (1986) and Allard (1984), with log K_d values of 5.3, 6.0, and 3.85 to 4.47 respectively. Zavarin et al (2004) proposes log K_d values ranging from 4.30 and 4.49 for Frenchman Flat and Pahute Mesa calcite systems respectively. Allard and Beall (1979) studied sorption of americium on dolomite and reported a log K_d value of 2.46 at pH 7.2 to 8.5. Dosch (1979), and Dosch and

Lynch (1980) reported log K_d ranges of 3.41 to 4.34, and 3.3 to 4.3 respectively in groundwater and brine solutions.

Curium

No data were available for curium transport in carbonate minerals, therefore average, minimum, maximum and standard deviation were not determined.

Table 2. Average, standard deviation, minimum, maximum and number of inputs for radionuclide log K_d in calcite, aragonate, limestone, dolomite and unidentified carbonate rocks.

	Calcite / Aragonite					**Limestone**					**Dolomite**					**Carbonate Rocks**				
Element	*Aver.*	*SD*	*Min*	*Max*	*N*	*Aver.*	*SD*	*Min*	*Max*	*N*	*Aver.*	*SD*	*Min*	*Max*	*N*	*Aver.*	*SD*	*Min*	*Max*	*N*
H	-	-	-	-	1	-	-	-	-	1	-	-	-	-	1	-	-	-	-	1
C	1.66	0.52	0.90	2.30	5	ND	ND	ND	ND	0	1.40	-	1.40	1.40	1	ND	ND	ND	ND	0
Cl	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Ar	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Ca	0.48	0.06	0.44	0.52	2	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Ni	-0.87	1.95	-2.24	0.51	2	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Kr	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Sr	-2.05	0.74	-3.00	-0.57	34	-1.84	1.17	-3.06	1.86	24	-1.99	0.79	-3.00	-0.74	10	-0.04	0.21	-0.19	0.11	2
Zr	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Nb	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Tc	ND	ND	ND	ND	0	-2.30	0.68	-3.05	-1.11	16	-2.96	0.70	-3.70	-1.82	5	-0.68	0.34	-0.92	-0.44	2
Pd	2.28	-	2.28	2.28	1	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Sn	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
I	ND	ND	ND	ND	0	-1.80	-	-1.80	-1.80	1	-3.00	ND	-3.00	-3.00	3	-0.14	0.03	-0.16	-0.12	2
Cs	-2.73	0.41	-3.57	-2.15	22	-0.94	1.09	-3.24	0.88	20	-1.16	1.12	-3.30	0.42	19	-0.96	0.79	-1.85	-0.36	3
Sm	3.46	1.44	1.04	4.67	5	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Eu	3.50	1.66	1.08	6.62	8	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Ho	3.06	-	3.06	3.06	1	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
Th	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0
U	-1.40	0.79	-3.07	-0.49	10	ND	ND	ND	ND	0	-2.00	0.11	-2.16	-1.92	4	0.33	0.18	0.20	0.45	2
Np(V)	0.94	0.94	0.08	3.70	46	ND	ND	ND	ND	0	-0.97	0.39	-1.54	-0.70	4	2.40	0.09	2.33	2.47	2
Pu(IV)	3.04	-	3.04	3.04	1	ND	ND	ND	ND	0	0	0	0	0	2	ND	ND	ND	ND	0
Pu(V)	1.96	0.62	1.30	2.74	4	ND	ND	ND	ND	0	-0.98	0.18	-1.19	-0.83	3	2.06	0.04	2.03	2.09	2
Am	2.37	1.04	1.46	6.00	42	ND	ND	ND	ND	0	3.56	0.78	2.46	4.34	5	ND	ND	ND	ND	0
Cm	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0	ND	ND	ND	ND	0

Table 3. Summary of available average, standard deviation, minimum, maximum and number of inputs for radionuclide K_d encompassing all carbonate minerals.

Element	Average	Min	Max	N
Hydrogen	-	-	-	1
Carbon	1.53	0.90	2.30	6
Calcium	0.48	0.44	0.52	2
Nickel	-0.87	-2.24	0.51	2
Strontium	-1.48	-3.57	1.86	70
Technetium	-1.98	-3.70	-0.44	23
Palladium	2.28	2.28	2.28	1
Iodine	-1.65	-3.00	-0.12	6
Cesium	-1.44	-3.57	0.88	64
Samarium	3.46	1.04	4.67	5
Europium	3.50	1.08	6.62	8
Holmium	3.06	3.06	3.06	1
Uranium	-1.02	-3.07	0.45	16
Neptunium (V)	0.79	-1.54	3.70	52
Plutonium (IV)	1.57	-0.11	3.04	3
Plutonium (V)	1.01	-1.19	2.74	9
Americium	2.97	1.46	6.00	47

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